Dual fiber-coupled laser hygrometer for fast in-situ gas analysis with minimized absorption path length

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ABSTRACT

Direct tunable diode laser absorption spectroscopy (dTDLAS) offers the possibility for highly selective, fast, calibration-free and sensitive in-situ measurements of individual gas components. Minimally invasive fiber-coupled sensors are especially interesting for numerous applications e.g. process or combustion analysis, where compact sensors for applications with limited optical access are required. Especially the sensor development for the gas analysis in internal combustion engines is challenging as harsh environmental conditions and very confined space have to be considered.

A new, very compact, high-speed diode laser hygrometer in the short-wave infrared with a fully fiber-coupled, minimally invasive sensor head was developed for highly localized absorption measurements in space confined applications like in engine diagnostics. The sensor optics has the dimensions of a typical engine spark plug, i.e. a small outer diameter of only 12 mm. To allow localized species measurements at the location of the ignition spark only a few millimeters absorption path are allowed for fast in-situ measurements of water vapor. To achieve a high sensitivity one major step in the spectrometer design is an application-specific absorption line selection in order to determine a target line with minimal spectral interference by other gas species, especially at higher pressures, optimized temperature dependence, as well as sufficient strong line strength. We isolated a well separated H_2O absorption line in the fundamental rovibrational band around the 2.6 µm region. A major problem of this spectral region is the lack of industrial tested optical components and materials, so that the component selection and design has a major influence on the spectrometer performance.

The design and development of the fiber-coupled dTDLAS hygrometer is discussed using customized IR optical components to realize a compact, minimally invasive dTDLAS sensor for harsh environments with a time resolution down to 100 µs. Laser, detector and electronic components are placed in one transceiver unit to be separated from the measurement point to reduce disturbances caused by vibrations as well as distortions of the results by ambient water vapour.

A sensor prototype was fabricated and a validation of the spectroscopic, optical and mechanical concept was successfully conducted. An optical resolution of $3 \cdot 10^{-3}$ absorbance was achieved. The sensor enables calibration-free H_2O measurements at 10 kHz with a detection limit of 0.06 Vol.-% with a theoretical dynamic range from 0.1 Vol.-% up to 100 Vol.-% at pressures from 0.1 – 0.5 MPa. Further, the uncertainties for water vapor measurements using the dTDLAS were significantly improved by measuring the required molecular parameter (e.g. absorption linestrength of the rovibrational transition) with high accuracy.

1. INTRODUCTION

The development of modern internal combustion (IC) engines is mainly driven by the need to increase fuel efficiency and minimize pollutant emission. A well-proven option to reduce engine NO_x emissions is exhaust gas recirculation (EGR). Here, parts of the burned combustion gases are scavenged inside the cylinder or returned to the combustion chamber [1]. Special attention must be paid to find an optimal adjustment of the recirculated gas fraction for various operating conditions. This requires quantification of the exhaust gas, especially at high EGR rates where inhomogeneous or fluctuating exhaust gas distributions can lead to misfire. Therefore, an in situ gas analysis directly inside the combustion chamber is needed. In situ measurements inside the engine impose high demands on the measuring technique. The physico-chemical parameters (pressure, temperature and gas matrix) rapidly change over a large dynamic range. The time before the ignition, i.e., the compression stroke is of central importance for EGR analysis. Here, temperatures between 300 and 600 K and pressures from around 0.1 MPa up to 1 MPa are prevailing, while the H_2O vapor concentration varies, depending on the EGR ratio, between 1 and 5 Vol.-%. In order to monitor EGR conditions, the temporal dynamics in the compression stroke should be resolved at typical engine speeds, i.e., 1,000–4,400 rpm. This requires a time resolution of the analytical system in the range of at least a few 100 µs.
The situation gets even more complicated in production engines that are not designed to provide large optical access to the combustion chamber. As every modification to production engines has the potential to change the combustion properties, minimally invasive optical interfaces based on fiber coupled endoscopic optics are desired, while optics must be robust, i.e., insensitive against window fouling and engine vibrations.

2. MEASUREMENT PRINCIPLE OF dTDLAS

dTDLAS uses a high spectral resolution, continuously tunable diode laser as light source [2]. The transmitted light is attenuated by wavelength-dependent molecule specific absorption, while the laser wavelength is periodically scanned by a triangular laser current modulation. The repetition rate of this modulation is chosen depending on the application requirements and is typically in the range between 100 Hz and 10 kHz. A time resolution in the ms to µs range is therefore possible and has already been demonstrated [3,4]. The transmission *T(ν)* can be described by the Lambert–Beer equation (1).

$$T(\nu) = \frac{I(\nu)}{I_0(\nu)} = e^{-S(T)\cdot\phi(\nu-\nu_0)\cdot N\cdot L} \quad (1)$$

This can be converted to extract the species volume mixing ratio, *c*, Eq. (2) by applying the ideal gas law:

$$c = \frac{-k_B\cdot T}{S(T)\cdot L\cdot p}\int ln\left(\frac{I(\nu)}{I_0(\nu)}\right)\frac{d\nu}{dt}\,dt. \quad (2)$$

Here *S(T)* denotes the temperature dependent molecular linestrength of the used H_2O transition, $\Phi(\nu-\nu_0)$ the line shape function, *N* the absorber number density, k_B the Boltzmann constant, *T* the measured gas temperature, *L* the absorption path length, $I(\nu)$ the measured intensity at the detector, the initial light intensity $I_0(\nu)$. Finally, *dν/dt* describes the dynamic wavelength tuning coefficient of the laser, which has to be determined experimentally. This is extracted from the Airy-signal when the laser light is transmitted through an etalon.

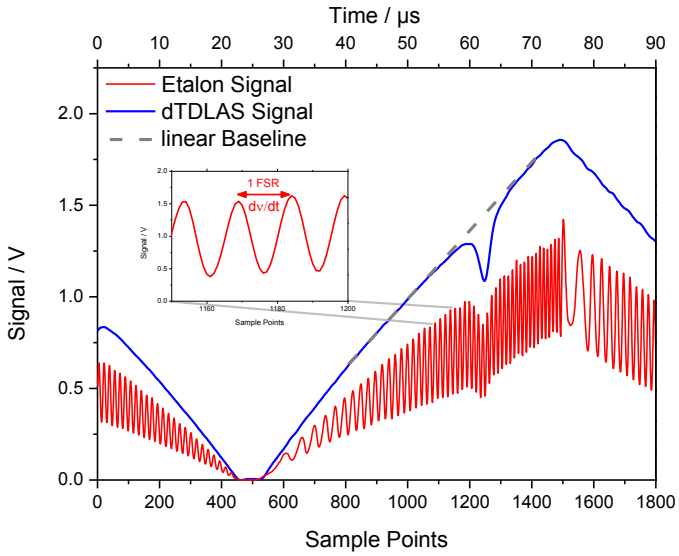

Figure 1: Time dependent TDLAS signal with the molecular absorption line and the corresponding signal through a Fabry-Pérot etalon.

Figure 1 shows the TDLAS signal during H_2O absorption and the corresponding signal through a 2.99 cm etalon. The peak to peak distance corresponds to the free spectral range (FSR) of the etalon which is defined as:

$$FSR = \frac{1}{2nd} \quad [cm^{-1}] \qquad (3)$$

Here n denotes the refractive index and d the etalon length in cm. In this case a solid silicon etalon with a FSR of $4{,}863 \cdot 10^{-2}$ cm^{-1} was used. Using the etalon as a relative wavenumber indicator time dependent TDLAS signal can be transferred to a linear wavenumber range. First an Airy fit is applied to the etalon signal to get the time dependent peak positions of the Fabry-Pérot interference. Subsequently a polynomial fit is used to the determined peak positions to retrieve the dynamic wavelength tuning coefficient *dv/dt* (Figure 2).

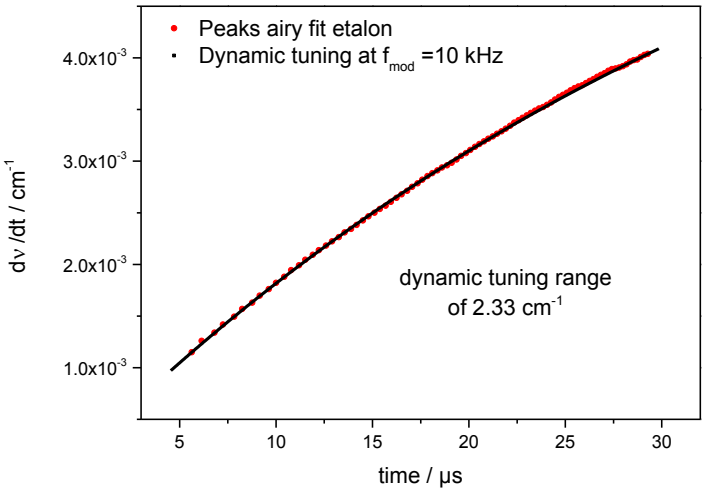

Figure 2: Polynomial fit to the time dependent maxima of the Fabry-Pérot interference to retrieve the dynamic wavelength tuning coefficient.

The linear base line of the TDLAS signal (Figure 1) has to be replaced by a polynomial baseline, due to the transformation of the time dependent TDLAS signal to a linear wavenumber range. Using a polynomial baseline fit the absorbance A ($A=\ln(I/I_0)$) of the ro-vibrational transition can be extracted and subsequently fitted using a Voigt line shape model as line shape function (Figure 3).

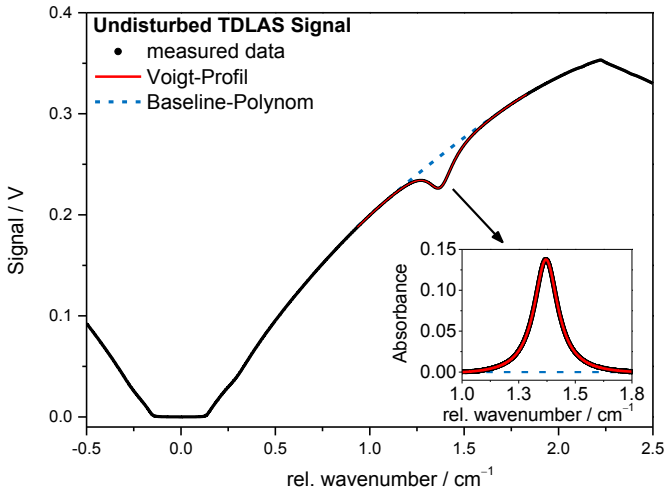

Figure 3: TDLAS signal in the linear rel. wavenumber range.

However, for most real world applications e.g. combustion analysis or airborne trace gas sensing, the signal is highly disturbed and needs to be corrected [5–9]. Signal disturbances may be caused by unwanted background emission *E(t)* of other radiation sources e.g. by flames or ambient light. Further the transmission *Tr(t)* fluctuates due to broadband light scattering on small particles. As this disturbances can be time dependent the modulation frequency of the laser are chosen to be fast enough to "freeze" the signal fluctuations over the time of a single TDLAS scan (*E(t)→E; Tr(t)→Tr*). Hence the modulation frequency has to be adjusted to the application to extract the information directly from the detector signal and subsequently correct the signal (Figure 4). The typical modulation frequency varies from 100 Hz for ambient on the trace gas sensing on the ground up to several kHz for applications in internal combustion engines. This correction employs an extended Lambert-Beer law.

$$c = \frac{-k_B \cdot T}{S \cdot L \cdot p} \int ln\left(\frac{I(\nu)-E}{I_0(\nu)\cdot Tr}\right)\frac{d\nu}{dt}dt. \qquad (4)$$

A closer look at Eq. (4) shows that no calibration parameter is needed to derive the absorber number density. Hence by simultaneously measuring pressure and temperature the absolute species concentration can be obtained without calibration against a spectrum- or concentration-reference, which is why we call this technique calibration-free. The absolute accuracy of this calibration-free technique has lately been demonstrated by a metrological comparison against a primary H_2O reference standard [10].

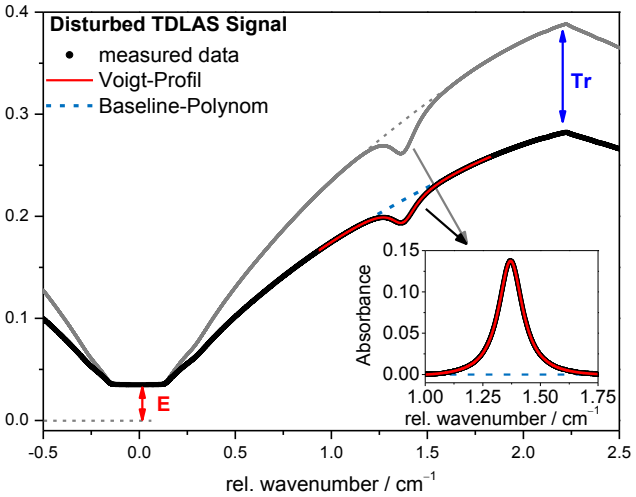

Figure 4: Emission and transmission correction of the TDLAS signal.

3. SELECTION OF THE ABSORPTION LINE AND LINESTRENGTH MEASUREMENT

For demanding applications such as EGR analysis in combustion engines special care must be taken to select a suitable absorption line. To quantify EGR, water vapor can be measured as a proxy to quantify the other gas components such as CO and CO_2 using an advanced simulation models for IC engines [11]. Figure 5 shows an absorption-line spectrum of the three combustion relevant gas species in the near infrared. Two absorption bands of H_2O are especially interesting. The 1.4 µm band offers highly developed optical components e.g. fibers, collimators and lasers, due to the development for the telecom industry. But, the absorption path length for minimal invasive sensors for IC engines is limited to 5 to 10 mm. Hence stronger absorption lines are required to ensure the spectrometer sensitivity. The 2.6 µm H_2O absorption band is up to 20 times stronger compared to the 1.4 µm band making minimal invasive sensors with small path lengths and sufficient sensitivity possible. However, the lack of standard products such as fibers, fiber-couplers and optical elements in this wavelength range on the other hand makes the design and fabrication of such sensors challenging.

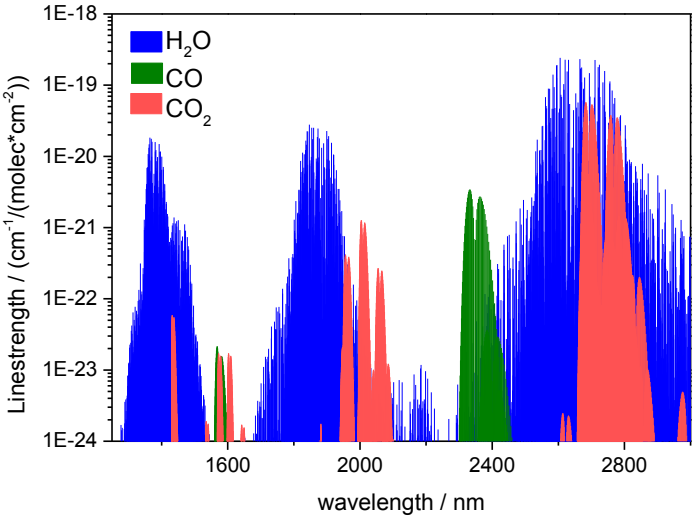

Figure 5: Absorption line spectrum of combustion relevant gas species. Molecular data from the HTRAN 2012 database is used.

An absorption line selection routine based on the HITRAN database was developed in our group and used to determine a target line [12,13]. The demanding boundary conditions such as minimal spectral interference by other gas species, especially at higher pressures, optimized temperature dependence as well as sufficient strong line were considered. The selected line 716 – 817 at 3920 cm^{-1} (2551 nm) offers the best compromise between line strength and isolation against other gas species as well as self interference with other H_2O absorption lines. The excellent line isolation minimizes the complexity of analysis at higher pressures. This is especially important as the wavelength tuning range of distributed feedback (DFB) laser rapidly decreases with higher modulation frequency (Figure 6). The wavelength tuning range decreases by about 50 % from 140 Hz to 10 kHz. The selected line leads to 5 % absorption at 1.5 Vol.-% H_2O with just a 10 mm absorption path length.

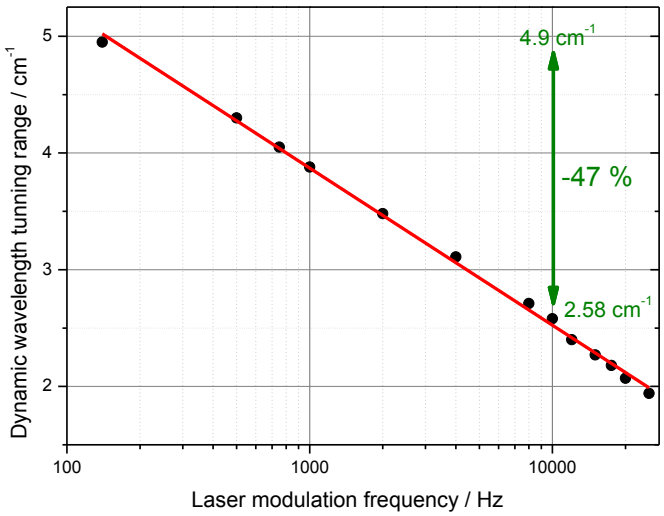

Figure 6: Measured frequency dependence of the tuning range for used DFB laser diode.

The line specific molecular parameter e.g. line strength and pressure broadening are very important for the fitting process and the accuracy of the laser spectrometer. Unfortunately the uncertainties of these molecular parameters given in the HITRAN database can range from 1 % up to 100 % (1σ). Looking at Eq. 3 this directly affects the overall concentration measurement. Therefore the quantitative accurate determination of the specific parameters is essential to reduce the uncertainties for the spectrometer. For the selected absorption line HITRAN states a line strength uncertainty of 10 %. Using a dTDLAS spectrometer with highly stable and known boundary conditions the line strength could be determined to $S = 2.57\ 10^{-20} \pm 2.35$ % (1σ) with very good reproducibility (Figure 7).The line strength is in very good agreement with other measurements reported in literature as well as with HITRAN but with considerable lower uncertainty. This strength measurement of the selected absorption line reduces the overall uncertainty for lab based H_2O concentration measurements from 10 % to 3.1 %.

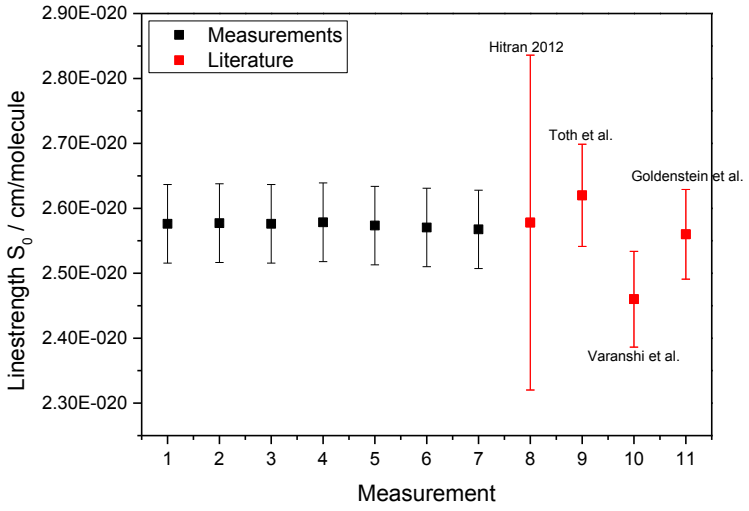

Figure 7: Line strength measurement of the 716-817 rovibrational transition at 3920.08 cm^{-1} of H_2O compared to literature data. The measurement shows a considerable lower uncertainty than the HITRAN database.

4. SENSOR DEVELOPMENT AND SETUP

The overall setup of a laser hygrometer is displayed in figure 8. The custom made DFB laser (Nanoplus) emitting at 2.551 µm is current modulated and temperature stabilized using a function generator (Agilent) and a laser driver (SRS). The current modulation introduces a wavelength shift to scan over the selected absorption line. The transmitted power is monitored using a photodiode. The photocurrent is converted in a voltage signal and amplified by a transimpedance amplifier (Femto) and subsequently digitized. The digitized signal is then used to retrieve the gas species concentration by the formalism described in the section 2.
The fast temperature and pressure variations in IC engines cause great demands on the time resolution of the spectrometer. A sampling of the absorption line has to be faster than variations of pressure and temperature to assure a reliable concentration measurement. This can be achieved by a scanning frequency of 10 kHz for most cases. This corresponds to a crank angle resolution of 1.2°CA at 2000 round per minute. To assure an uninterrupted measurement a high speed data acquisition system was built using 33 MS/s with a 14 bit resolution.

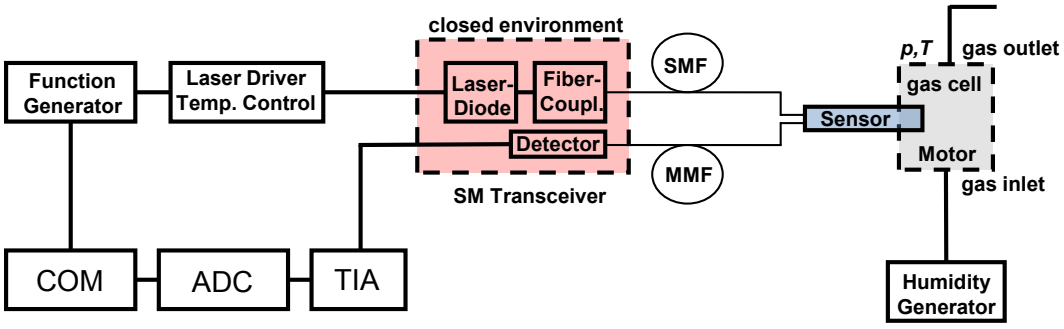

Figure 8: Setup of the laser hygrometer. The minimal invasive sensor head is decoupled from the DFB diode laser and the receiver electronic using a special designed fiber coupling.

A fiber coupling interface is essential for a 2.5 µm minimal invasive laser hygrometer to decouple the transceiver unit from the IC engine. This is necessary to avoid optical and electronic noise caused by the vibration of the engine. Further the influence of unwanted absorption due to ambient air humidity has to be minimized. This unwanted "parasitic" absorption would decrease the sensor sensitivity especially with small absorption path length of the sensor of 21 mm [14]. Therefore a special optic made of zinc selenide were designed and fabricated to enable a collimation and coupling of the strongly divergent diode laser beam. The elliptical beam profile is coupled into a ZBLAN single mode fluoride fiber (SMF) to avoid free space paths in ambient air. Laser, lenses and fiber coupling are placed in a separate closed transceiver unit which can be purged by N_2 or dry air. Self made fiber-feedthroughs guide the radiation out an into transceiver unit. A SMF guides the radiation to the sensor head, where it is transmitted through the combustion chamber using a lens (Figure 9). After absorption in the gas phase the radiation is collected by the lens and a multimode fiber (MMF, ZBLAN) and guided back to the detector placed in the purged transceiver unit.

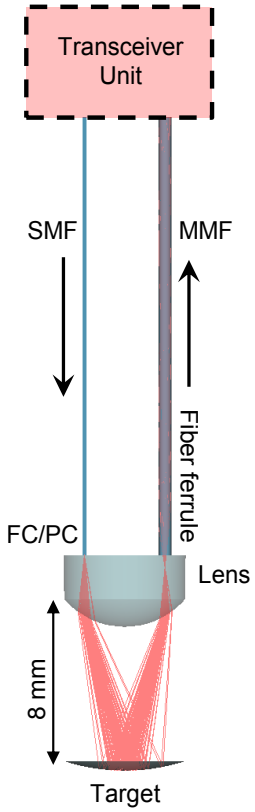

Figure 9: Setup of the minimal invasive sensor head

A full size prototype was built to validate the optical concept of the sensor head. The lens, SMF and MMF were built into a threaded cylinder with a diameter of 12 mm (M12). The lens acts as a transfer optical element to guide the light from the SMF to the target element. The target reflects/scatters the light back were the collected light is focused on the MMF (600 µm core). The sensor head was pressure tight capsule to allow a validation under of local H_2O concentration measurement under engine like pressure conditions.

5. SENSOR VALIDATION AND RESULTS

The sensor head was screwed into a measurement chamber to validate the spectrometer (Figure 8). H_2O concentrations between 0.1 Vol.-% and 1 Vol.-% were provided by a humidity generator (Thunder Scientific) to test the linearity of the laser hygrometer. Figure 10 displays the results of the absolute concentration measurement by the spectrometer using dTDLAS in comparison to the generated concentration steps. A very good linearity of the laser hygrometer could be achieved resulting in a slope of 0.999. It should be emphasized that no signal averaging or filtering was applied to reduce electronic noise due to the required high time resolution. Despite these difficult boundary conditions a signal to noise ratio (SNR) of 40 at 2.3 Vol.-% was achieved resulting in a detection limit of 0.058 Vol.-%. This corresponds to an optical resolution of $3.6 \cdot 10^{-3}$ absorbance, where SNR is defined as the standard deviation (1σ) of the residual between measured and fitted line profile. The precision of the spectrometer including the minimal invasive sensor head is 160 ppm measured at 1 Vol.-% H_2O.

The sensor head was quite insensitive to vibration during the H_2O measurement, which is essential for IC engine measurements. The settings of the spectrometer were adjusted towards real engine conditions. The laser modulation frequency was 10 kHz, which leads to a wavelength tuning range of 2.33 cm^{-1} (Figure 6). The loss in tuning depth limits the analysis of higher pressure signals as not the entire absorption line can be scanned at high pressures.

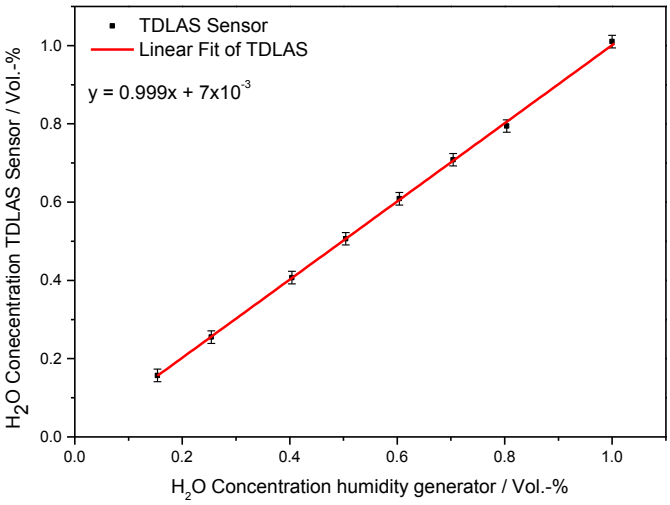

Figure 10: Measured linearity of the Laser hygrometer. The very good linearity results in a slope of 0.999 for the linear regression.

Figure 11 shows the non averaged line profiles with fitted Voigt line shapes measured by the fiber-coupled minimal invasive laser hygrometer. The residual, the difference between measurement and fit, is displayed in the lower part of each scan. The residual standard deviation relates to the sensitivity of the spectrometer. Pressurized air was saturated at constant temperature and the pressure and thus the H_2O-concentration was varied to investigate pressures from 0.1 MPa (2.3 Vol.-%) up to 0.5 MPa (0.47 Vol.-%). Under these conditions SNR of 40 (2.3 Vol.-%, 0.1 MPa) and 12 (0.47 Vol.-%, 0.5 MPa) were achieved, respectively.

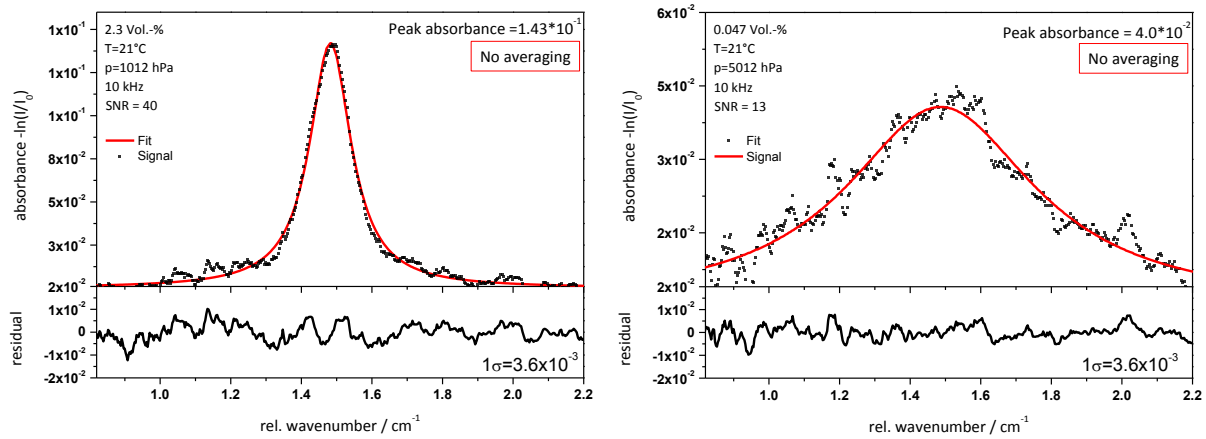

Figure 11: Single scans of H_2O measurements using the minimal invasive sensor with the selected absorption line at 2.55 µm with 10 kHz modulation frequency. No scan averaging was performed to achieve a time resolution of 100 µs. The measurement was conducted under different pressure ranging from 0.1 MPa to 0.5 MPa at 21°C.

The metrological uncertainties of the laser hygrometer have been analyzed according to GUM [15]. The uncertainty budget is shown in table 1. For lab based measurements the linestrength (48.2 %) and the line area (34.9 %) show the biggest influence on the total uncertainty of 3.1 % (1σ). This reveals the high importance on uncertainty quantification of molecular parameter such as line strength as well as foreign and self broadening coefficient which are important for the Voigt profile fitting routine to retrieve the line area. However, it should be

mentioned that these uncertainties not necessarily represent the values for IC engine measurements as the uncertainty of the temperature and pressure in this environments are higher.

Table 1: Uncertainty budget of laboratory measurements

Quantity	Name	Value	Uncertainty	Influence
T	Temperature	294 K	0.2 K	0 %
$S(T_0)$	Linestrength (T_0 = 296 K)	$2.58 \cdot 10^{-20}$ cm^{-1} (molec·cm^{-2})	2.35 %	**48.2 %**
p	Pressure	1012 mbar	2 mbar	0.1 %
L	Absorption path length	21 mm	0.5 mm	16.5%
A	Line area	0.0318 cm^{-1}	2 %	**34.9 %**
Results				
c_{H2O}	**H_2O-concentration**	**(2.38 ± 0.08) Vol.-%**	**3.4 %**	–

6. CONCLUSION AND OUTLOOK

A suitable H_2O absorption line was selected using criteria necessary for the boundary conditions in IC engines. Molecular parameters of the selected line were measured to improve the uncertainty from 10 % to 3.4 for the developed laser hygrometer.
A prototype of a minimally invasive dTDLAS hygrometer for in-situ measurements of absolute H_2O-concentrations in 2.5 µm range was successfully developed. A fiber coupled screw-in sensor using IR-MIR fluoride fibers offers the possibility to quantify local residual gas in IC engines with just one access point. The developed hygrometer reaches a time resolution of up to 100 µs which corresponds to a potential crank angle resolution of 1.2°CA at 2000 rpm. The sensor showed a detection limit of 0.06 Vol.-% H_2O over a calculated dynamic range from 0.1 to 100 Vol.-%. Where an absorbance of 1 (66 % absorption) corresponds to 100 Vol.-% H_2O. The sensor was validated up to 0.5 MPa. The optical concept seems therefore suitable for a fiber-coupled dTDLAS hygrometer for IC engines applications.
Currently the sensor is prepared for first test measurements in an optical research engine. Further the use of vertical cavity emitting laser (VCSEL) is proofed which could extend the measurements to higher pressures. The sensor transmission from 0.4 µm to 4 µm offers the possibility to detect other combustion relevant gas species such as CO, CO_2 or O_2 by carefully chosen laser wavelength.

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